

# Exact double-counting in combining the Dynamical Mean Field Theory and the Density Functional Theory

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We propose a continuum representation of the Dynamical Mean Field Theory, in which we were able to derive an exact overlap between the Dynamical Mean Field Theory and band structure methods, such as the Density Functional Theory. The implementation of this exact double-counting shows improved agreement between theory and experiment in several correlated solids, such as the transition metal oxides and lanthanides. Previously introduced nominal double-counting is in much better agreement with the exact double-counting than most widely used fully localized limit formula.

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Understanding the electronic structure of materials with strong electronic correlations remains one of the great challenges of modern condensed matter physics. The first step towards calculating the electronic structure of solids has been achieved by obtaining the single-particle band dispersion  $E(\mathbf{k})$  within the density functional theory (DFT) in the local density approximation (LDA) [1], which takes into account correlation effects only in a limited extent.

To account for the many-body correlation effects beyond LDA, more sophisticated methods have been developed. Among them, one of the most successful schemes is the dynamical mean-field theory (DMFT) [2]. It replaces the problem of describing correlation effects in a periodic lattice by a strongly interacting impurity coupled to a self-consistent bath [3]. This method was first developed to solve the Hubbard model, but it was soon realized [4] that it can also be combined with the LDA method, to give more material specific predictions of correlation effects in solids. The LDA+DMFT method achieved great success in the past two decades, as it was successfully applied to numerous correlated solids [5]. The combination of the two methods, nevertheless lead to a problem of somewhat ambiguous way of subtracting the part of correlations which are accounted for by both methods.

The so-called double-counting (DC) term was usually approximated by the formula first developed in the context of LDA+U, and was evaluated by taking the atomic limit for the Hubbard interaction term [6, 7]. Many other similar schemes were proposed recently [8–11], but rigorous derivation of this double-counted interaction within DMFT and LDA is missing to date. Here we propose a new method of calculating the overlap between DMFT and a band-structure method (either DFT or GW), and we explicitly evaluate this DC functional within LDA+DMFT applied to well studied transition metal oxides such as  $\text{SrVO}_3$ ,  $\text{LaVO}_3$ , and most studied lanthanide metal, the elemental Cerium.

To compare different approximations in the same language, it is useful to cast them into the form of the Luttinger Ward functional [5, 12, 13], which is a func-

tional of the electron Green's function  $G$ , and takes the form  $\Gamma[G] = -\text{Tr}((G_0^{-1} - G^{-1})G) + \text{Tr} \log(-G) + \Phi_{V_c}[G]$ . The first part is the material dependent part, in which  $G_0^{-1}(\mathbf{r}\mathbf{r}'; \omega) = [\omega + \mu + \nabla^2 - V_{ext}(\mathbf{r})]\delta(\mathbf{r} - \mathbf{r}')$ , and the second two terms are universal functionals of the Green's function  $G(\mathbf{r}\tau, \mathbf{r}'\tau')$  and the Coulomb interaction  $V_c(\mathbf{r} - \mathbf{r}')$ . In the exact theory,  $\Phi_{V_c}[G]$  contains all skeleton Feynman diagram, constructed by  $G$  and  $V_c$  [13]. In the language of Luttinger Ward functional, different approximations can then be looked at as different approximations to the interacting part  $\Phi_{V_c}[G]$ .

The Density Functional Theory can be derived by approximating the exact functional  $\Phi_{V_c}[G]$  by  $E_H[\rho(\mathbf{r})] + E_{xc}[\rho(\mathbf{r})]$ , where  $E_H$  and  $E_{xc}$  are the Hartree and the exchange-correlation functionals, respectively. The stationarity condition gives the DFT equations, i.e.,  $G^{-1} - G_0^{-1} = (V_H[\rho] + V_{xc}[\rho])\delta(\mathbf{r} - \mathbf{r}')\delta(\tau - \tau')$ , because  $\delta E_{xc}[\rho]/\delta G = \delta(\mathbf{r} - \mathbf{r}')\delta(\tau - \tau') \delta E_{xc}[\rho]/\delta \rho = \delta(\mathbf{r} - \mathbf{r}')\delta(\tau - \tau') V_{xc}[\rho]$ . Note that in this language, exact DFT appears as an approximation to the exact Green's function, where the exact self-energy is approximated by a static and local potential. Note also that the static approximation is a consequence of truncating the variable of interest, namely replacing full  $G(\mathbf{r}, \tau, \mathbf{r}', \tau')$  by its diagonal components  $\rho(\mathbf{r}) = \delta(\mathbf{r} - \mathbf{r}')\delta(\tau - \tau')G(\mathbf{r}\tau, \mathbf{r}'\tau')$ .

In the Luttinger-Ward functional language, the DMFT appears as an approximation where the Green's function in the  $\Phi$  functional is replaced by its local counterpart  $G \rightarrow G_{local}$ , and the Coulomb repulsion  $V_c$  by screened interaction  $V_c \rightarrow U$ , namely  $\Phi^{DMFT} = \Phi_U[G_{local}]$ . [5] Note that the DMFT functional has exactly the same form as the exact functional  $\Phi_{V_c}[G]$ , because all the skeleton Feynman diagrams constructed by  $G_{local}$  and  $U$  are summed up by DMFT, while in DFT the functional  $E_{xc}[\rho]$  is unknown, and further approximation is necessary. The truncation of the variable of interest from  $G$  to  $G_{local}$  leads in DMFT to self-energy, which is also local in space, but it keeps its dynamic nature. Other approximations such as Hartree-Fock or GW can be similarly derived by replacing  $\Phi_{V_c}[G]$  by some limited set of Feynman diagrams, i.e., truncation in space of Feyn-

man diagrams, rather than truncation of the variable of interest.

There is some kind of disconnect between the DMFT functional  $\Phi_U^{DMFT}[G_{local}]$ , and the LDA functional  $E_{xc}[\rho(\mathbf{r})]$ , mostly because the auxiliary systems for the two methods are very different. The auxiliary system for LDA approximation is the uniform electron gas problem defined for continuum, in the absence of complexity of the solid. On the other hand, DMFT is usually associated with the lattice model like Hubbard model, where mapping to the local problem reduces to the Anderson impurity model, which does not have a well-defined continuum representation. The double-counting problem occurs because it is not clear what is the overlap between the two methods, i.e., what physical processes are accounted for in one and what in the other method.

It is useful to represent the DMFT method in the continuum representation. Such representation is not unique, but physical intuition can guide the mapping. Here we propose to look at the DMFT problem as the approximation, which solves exactly the problem defined by some auxiliary Green's function  $G_{local} = \hat{P}G$  and Coulomb repulsion replaced by Yukawa short-range interaction  $V_c^\lambda = \frac{e^{-\lambda|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|}$ . We have in mind some projector  $\hat{P}$ , which is very local, and truncates the Green's function to a region mostly concentrated inside the muffin-tin sphere. It can for example be defined by a set of quasi-atomic orbitals  $G_{local}(\mathbf{r}, \mathbf{r}') = \sum_{L,L'} \langle \mathbf{r} | \Phi_L \rangle \langle \Phi_L | G | \phi_{L'} \rangle \langle \phi_{L'} | \mathbf{r}' \rangle$  where  $\langle \mathbf{r} | \Phi_L \rangle = u_L(r) Y_L(\mathbf{r})$  are spheric harmonics times localized radial wave function. Note that this truncation of the Green's function  $G(\mathbf{r}, \mathbf{r}')$  to its local counterpart parallels the truncation of the Green's function to its diagonal component in theories that choose density as the essential variable, i.e.,  $\rho(\mathbf{r}) = G(\mathbf{r}\tau, \mathbf{r}'\tau')\delta(\mathbf{r}-\mathbf{r}')\delta(\tau-\tau')$ . The screening  $\lambda$  in Yukawa interaction  $V_c^\lambda$  has to be large enough such that the interaction between electrons on neighboring sites is negligible. The DMFT can then give an exact Luttinger-Ward functional  $\Phi_{V_c^\lambda}[\hat{P}G]$ , i.e., containing all local Feynman diagrams constructed by  $\hat{P}G$  and  $V_c^\lambda$ , defined in the continuum [14]. The stationarity condition for the Luttinger-Ward functional gives the DMFT equations  $G^{-1} - G_0^{-1} = \hat{P} (\delta\Phi_{V_c^\lambda}[G_{local}]/\delta G_{local})$ .

The precise determination of the screening  $\lambda$  is beyond the scope of this paper. However, we notice that once the Coulomb interaction  $U$  in DMFT is known, the screening length  $\lambda$  is uniquely determined by  $U$  through the matrix elements of the Yukawa interaction in DMFT basis. Notice that Hund's coupling  $J$  is not a free parameter in this parametrization, but is uniquely determined by  $\lambda$  through Yukawa form of the Coulomb interaction. [24]

After the mapping of the DMFT method to the continuous  $(\mathbf{r}, \mathbf{r}')$  Hilbert space, where DFT exchange-correlation is defined, it is easy to see what is the

overlap between the two methods. The Hartree term is accounted for exactly in the LDA method, and has the form  $E_{V_c}^H[\rho] = \frac{1}{2} \int d\mathbf{r} d\mathbf{r}' \rho(\mathbf{r}) \rho(\mathbf{r}') V_c(\mathbf{r} - \mathbf{r}')$ , while in DMFT it takes the following form  $E^{H,DMFT} = \frac{1}{2} \int d\mathbf{r} d\mathbf{r}' (\hat{P}\rho(\mathbf{r})) (\hat{P}\rho(\mathbf{r}')) V_c^\lambda(\mathbf{r} - \mathbf{r}')$ , which can also be written as  $E^{H,DMFT} = E_{V_c^\lambda}^H[\hat{P}\rho]$ , where  $\hat{P}\rho = \delta(\mathbf{r} - \mathbf{r}')\delta(\tau - \tau')G_{local}(\mathbf{r}\tau, \mathbf{r}'\tau') = \delta(\mathbf{r} - \mathbf{r}')\delta(\tau - \tau')\hat{P}G(\mathbf{r}\tau, \mathbf{r}'\tau')$ , and  $E_{V_c}^H[\rho]$  is the exact Hartree functional defined above. The Hartree contribution to the DC within LDA+DMFT (or any other band structure method which includes exact Hartree term) is thus  $E_{V_c^\lambda}^H[\hat{P}\rho]$  [15]. This DC term thus corresponds to truncating Green's function  $G$  and Coulomb interaction  $V_c$  by their local counterparts, i.e.,  $G \rightarrow \hat{P}G$  and  $V_c \rightarrow V_c^\lambda$ .

For approximations, which truncate in the space of Feynman diagrams (such as Hartree-Fock or GW method), one can obtain the DMFT double-counting by applying both the truncation in space of Feynman diagrams as well as the DMFT truncation in variables of interest. For the case of GW method, one can check diagram by diagram that the corresponding DMFT Feynman diagram is obtained by replacing  $G$  by  $\hat{P}G$  and  $V_c$  by  $V_c^\lambda$  in each diagram, just like it was done above for the Hartree term. More precisely, the GW functional can be written as  $\Phi_{V_c}^{GW}[G] = E_{V_c}^H - \frac{1}{2} \text{Tr} \log(1 - V_c G * G)$ , where  $G * G = P$  is the convolution of two Green's functions (polarization function). The GW+DMFT double-counting is thus  $E^{H,DMFT} - \frac{1}{2} \text{Tr} \log(1 - V_c^\lambda(\hat{P}G) * (\hat{P}G))$ , which can be shortly written as  $\Phi_{V_c^\lambda}^{GW}[\hat{P}G]$ .

In the case of DFT+DMFT, the expansion in terms of Feynman diagrams is not possible, however, to identify the overlap between the two methods, this is not essential. Clearly, the double-counting in DFT+DMFT is obtained by the same procedure of replacing  $G$  by  $\hat{P}G$  and  $V_c$  by  $V_c^\lambda$  in the DFT functional. Since the DFT also truncates the Green's function to its diagonal components only ( $\rho = \delta(\tau - \tau')\delta(\mathbf{r} - \mathbf{r}')G$ ) the DC is a functional of the local charge only  $\rho_{local} = \hat{P}\rho$ . DC thus takes the form

$$\Phi_{DC}^{DFT+DMFT} = E_{V_c^\lambda}^H[\hat{P}\rho] + E_{V_c^\lambda}^{XC}[\hat{P}\rho].$$

In LDA method, the exchange-correlation functional is obtained from the energy of the uniform electron gas. To obtain the LDA+DMFT double-counting, one thus needs to solve the problem of the electron gas with the density that contains only "local" charge  $\hat{P}\rho$ , which interacts by the screened Yukawa interaction  $V_c^\lambda$ . [24]

Including the exact double-counting, the LDA+DMFT  $\Phi$  functional is thus

$$\Phi^{LDA+DMFT}[G] = E_{V_c}^H[\rho] + E_{V_c}^{XC}[\rho] + \Phi_{V_c^\lambda}[\hat{P}G] - E_{V_c^\lambda}^H[\hat{P}\rho] - E_{V_c^\lambda}^{XC}[\hat{P}\rho], \quad (1)$$

where  $\Phi_{V_c^\lambda}[\hat{P}G]$  is the DMFT functional which contains all Feynman diagrams constructed from  $\hat{P}G$  and  $V_c^\lambda$ .

This is the central equation of this paper, as it defines the LDA+DMFT approximation including the exact DC. The saddle point equations give the LDA+DMFT set of equations

$$G^{-1} - G_0^{-1} = \hat{P} \frac{\delta \Phi_{V_c^\lambda}[G_{local}]}{\delta G_{local}} + \left( \frac{\delta E_{V_c^\lambda}^{HXC}[\rho]}{\delta \rho} - \hat{P} \frac{\delta E_{V_c^\lambda}^{HXC}[\rho_{local}]}{\delta \rho_{local}} \right) \delta(\mathbf{r} - \mathbf{r}') \delta(\tau - \tau') \quad (2)$$

where we used  $E^{HXC}[\rho] \equiv E^H[\rho] + E^{XC}[\rho]$  and  $\hat{P}G \equiv G_{local}$ .

The only difference between functional Eq. 1, and the usual LDA+DMFT implementation, is the presence of  $E_{V_c^\lambda}^{HXC}$ . This is the semi-local exchange and LDA correlation functional of the electron gas interacting by Yukawa interaction. The semi-local exchange-density  $\varepsilon_{V_c^\lambda}^x[\rho]$  (defined by  $E^x[\rho] = \int d\mathbf{r} \rho(\mathbf{r}) \varepsilon^x[\rho(\mathbf{r})]$ ), can be computed analytically, and takes the following form

$$\varepsilon_{V_c^\lambda}^x[\rho] = -\frac{C}{r_s} f(x)$$

where

$$f(x) = 1 - \frac{1}{6x^2} - \frac{4 \arctan(2x)}{3x} + \frac{(12x^2 + 1) \log(1 + 4x^2)}{24x^4},$$

$$C = \frac{3}{2} \left( \frac{9}{4\pi^2} \right)^{1/3}, \quad r_s = \left( \frac{3}{4\pi\rho} \right)^{1/3}, \quad \text{and} \quad x = \left( \frac{9\pi}{4} \right)^{1/3} \frac{1}{\lambda r_s}.$$

The exchange potential  $V^x = \frac{\delta}{\delta \rho} E^x[\rho]$  is then  $V_{V_c^\lambda}^x = \frac{4}{3} \varepsilon_{V_c^\lambda}^x + \frac{1}{3} \frac{C}{r_s} x \frac{df}{dx}$ .

The correlation part requires solution of the homogeneous electron gas problem interacting with Yukawa repulsion, which was solved by QMC [16–18]. Here we want to have an analytic expression for correlation energy at arbitrary  $\lambda$  and  $r_s$ . It is well established that  $G_0W_0$  gives quite accurate correlation energy of the electron gas [19, 20], especially when computed from the Luttinger-Ward functional  $\Gamma[G]$ . We thus repeated  $G_0W_0$  calculation for the electron gas, but here we use Yukawa interaction. We evaluate the total energy using Luttinger-Ward functional of GW to achieve high accuracy. We then fit the correlation energy in the range of physically most relevant  $r_s \in [0, 10]$  and screenings  $\lambda \in [0, 3]$  ( $\lambda$  is measured in Bohr radius inverse) with the following functional form:

$$\varepsilon_{V_c^\lambda}^c = \frac{\varepsilon_{\lambda=0}^c}{1 + \sum_{n=1}^4 a_n r_s^n} \quad (3)$$

Ce- $\alpha$	$n_f$	$V_{dc}/U$
exact	0.997	0.424
nominal	1.002	0.500
FLL	1.035	0.533

TABLE I: LDA+DMFT valence and DC potential for  $\alpha$ -Ce at  $T = 200$  K. The local Coulomb repulsion in Ce is  $U = 6$  eV.

where

$$\log(1 + a_1) = \frac{\lambda(\alpha_0 + \alpha_1 \lambda)}{1 + \alpha_2 \lambda^2 + \alpha_3 \lambda^4 + \alpha_4 \lambda^6} \quad (4)$$

$$\log(1 + a_2) = \frac{\lambda^2(\beta_0 + \beta_1 \lambda)}{1 + \beta_2 \lambda^2 + \beta_3 \lambda^4} \quad (5)$$

$$\log(1 + a_3) = \frac{\lambda^3(\gamma_0 + \gamma_1 \lambda)}{1 + \gamma_2 \lambda^2} \quad (6)$$

$$\log(1 + a_4) = \lambda^4(\delta_0 + \delta_1 \lambda^2) \quad (7)$$

The best fit gives the following coefficients:

$$\begin{aligned} \alpha_i &= [1.2238912, 7.3648662, 9.6044695, \\ &\quad -0.7501634, 0.0207808] \times 10^{-1} \\ \beta_i &= [5.839362, 11.969474, 10.156124, 1.594125] \times 10^{-2} \\ \gamma_i &= [8.27519, 5.57133, 17.25079] \times 10^{-3} \\ \delta_i &= [5.29134419, 0.0449628225] \times 10^{-4} \end{aligned} \quad (8)$$

Finally, the correlation potential is  $V_\lambda^c = \frac{V_{\lambda=0}^c}{A(r_s, \lambda)} + \frac{\varepsilon_{\lambda=0}^c}{C(r_s, \lambda)}$ , where  $A(r_s, \lambda) = 1 + \sum_{n=1}^4 a_n r_s^n$  and  $C(r_s, \lambda) = 3[1 + \sum_{n=1}^4 a_n r_s^n]^2 / \sum_{n=1}^4 n a_n r_s^n$ . We take the unscreened correlation energy density  $\varepsilon_{\lambda=0}^c$  (and unscreened potential) from the standard parametrization of quantum Monte Carlo results, hence the  $G_0W_0$  calculation is only used for renormalization of correlations by screening with Yukawa form.

In the following we present results for some of the most often studied correlated solids, namely, elemental Cerium,  $\text{SrVO}_3$  and  $\text{LaVO}_3$ . We used three different forms of DC functional: i) "exact", which we introduced above, ii) "FLL" stands for fully localized limit form introduced in Ref. 6, which has the simple form  $V_{dc} = U(n - 1/2) - J/2(n - 1)$ , and  $n$  stands for the correlated occupancy, c) and the "nominal" DC, introduced in Ref. 9, 10. The "nominal"  $V_{dc}$  takes the same form as "FLL" formula, but  $n$  in the formula is replaced by the closest integer value ( $n^0 = [n]$ ), and hence  $n^0$  corresponds to so-called nominal valence. We use LDA+DMFT implementation of Ref. 9.

The physical properties of correlated materials are very sensitive to the value of the local occupancy  $n_f$ , and  $n_f$  is sensitive to the value of DC. In table I we show results for elemental Cerium in the  $\alpha$  phase. All three DC functionals give very similar correlated occupancies  $n_f$ , and all are very close to nominal valence

SrVO <sub>3</sub>	$n_{t2g+eg}$	$n_{t2g}$	$n_{eg}$	$V_{dc}^{t2g}/U$	$V_{dc}^{eg}/U$
exact	2.223	1.507	0.716	1.384	1.406
nominal	2.251	1.541	0.710	1.443	1.444
FLL	2.529	1.699	0.830	1.943	1.943

TABLE II: LDA+DMFT results for SrVO<sub>3</sub> at  $T = 200$  K and  $U = 10$  eV. Both  $t2g$  and  $eg$  orbitals are treated by DMFT.

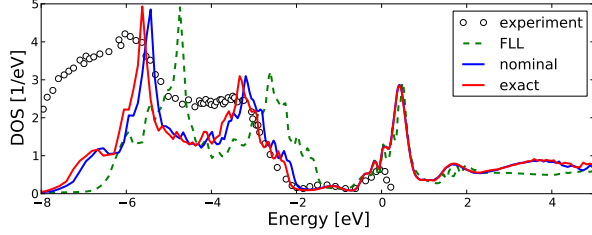


FIG. 1: (Color online) LDA+DMFT total density of states for SrVO<sub>3</sub> using three different DC potentials. Experimental photoemission is reproduced from Ref. 22. (parameters listed in table II).

$n^0 = 1$ . The actual value of the DC potential  $V_{dc}$  differs for less than  $0.1U$ , which leads to almost indistinguishable spectra on the real axis, and from the previously published results [9], hence we do not reproduce them here. We found a general trend in all materials studied that the exact DC is somewhat smaller than given by FLL formula. For Ce, the Hartree contribution to DC potential is  $V_H = n_f U \approx 0.997U$ , the semi-local exchange contribution is  $V_x \approx -0.485U$  and LDA correlation is  $V_c \approx -0.088U$ , hence the total DC potential is  $V_H + V_x + V_c \approx 0.424U$ , which is slightly smaller than FLL formula  $U(n_f - 1/2) - J/2(n_f - 1) \approx 0.533U$  or nominal formula  $U(n_f^0 - 1/2) - J/2(n_f^0 - 1) = 0.5U$ . It is interesting to note that the semi-local exchange used in LDA is quite different from the exact exchange value. The latter is only  $|V_F| = Un/14 \approx 0.071U$ , a substantially smaller value than the semi-local exchange  $|V_x| \approx 0.485U$ . This shows why DC within LDA+DMFT is so different from the Hartree-Fock value of the DMFT self-energy, i.e.,  $\Sigma(\omega = \infty)$ .

Next we present tests for SrVO<sub>3</sub>, which is a metallic transition metal oxide with nominally single electron in the  $t2g$  shell. Near the Fermi level  $E_F$ , there are mostly  $t2g$  states. The majority of  $eg$  states are above  $E_F$ , however, due to strong hybridization with oxygen some part of  $eg$  orbitals also gets filled. There are two ways the DMFT method can be used here. In the first case, one can treat only the  $t2g$  shell within DMFT. The vast majority of DMFT calculations for SrVO<sub>3</sub> were done in this way. In this case, all three DC potentials again give very similar results and the spectra is almost indistinguishable from previously published results in Ref. 10. One can also treat dynamically with DMFT the entire  $d$  shell. This

LaVO <sub>3</sub> ( $t2g$ -only)	$n_{t2g}$	$V_{dc}^{t2g}/U$	$V_{dc}^{eg}/U$
exact	2.014	1.195	1.193
nominal	2.074	1.450	1.450
FLL	2.099	1.544	1.544

TABLE III: LDA+DMFT results for LaVO<sub>3</sub> at  $T = 200$  K and  $U = 10$  eV. Only  $t2g$  orbitals are treated by DMFT.

LaVO <sub>3</sub> ( $t2g+eg$ )	$n_{t2g+eg}$	$n_{t2g}$	$n_{eg}$	$V_{dc}^{t2g}/U$	$V_{dc}^{eg}/U$	$V_{dc}^{eg}$
exact	2.444	2.048	0.397	1.596	1.599	1.665
nominal	2.344	2.032	0.312	1.458	1.458	1.458
FLL	2.706	2.167	0.540	2.114	2.114	2.114

TABLE IV: LDA+DMFT results for LaVO<sub>3</sub> at  $T = 200$  K and  $U = 10$  eV. Both  $t2g$  and  $eg$  orbitals are treated by DMFT.

case is presented in Table II and spectra in Fig. 1. One can notice that the exact and the nominal DC give very similar  $n_d$ , while the FLL formula gives 14% larger  $n_d$ . This is because the value of the DC potential is substantially larger ( $\approx 40\%$ ) when using FLL as compared to exact case. It is nevertheless comforting to see that 40% error in double-counting still does not lead to major failure of LDA+DMFT. We plot the spectra in Fig. 1, to show how this change in  $V_{dc}$  leads to shift of oxygen- $p$  spectra relative to vanadium- $d$  states. For the exact DC, the oxygen peak positions match very well with the experimentally measured spectra. The nominal valence is quite close to the exact spectra, while FLL formula leads to an upward shift of oxygen for roughly 0.6 eV, which is still relatively small compared to the difference in the double-counting potentials, which is  $V_{dc}^{FLL} - V_{dc}^{exact} \approx 5.37$  eV.

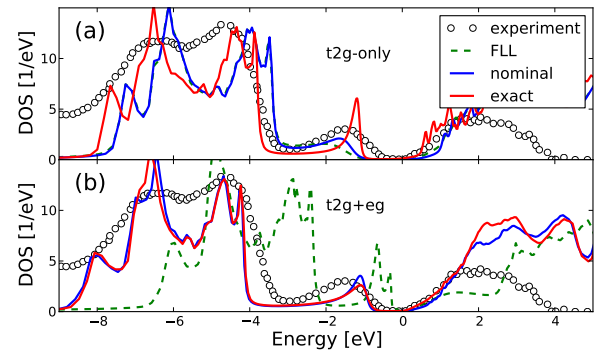


FIG. 2: (Color online) LDA+DMFT total density of states for LaVO<sub>3</sub> using the three different DC formulas. (a) only  $t2g$  orbitals are treated by DMFT (b) both  $t2g$  and  $eg$  orbitals are treated dynamically. Experimental photoemission is reproduced from Ref. 23.

Next we present results for the Mott insulating oxide LaVO<sub>3</sub>, which is solved in two ways, i) treating only

the  $t2g$  orbitals dynamically with DMFT, presented in Table III and Fig. 2a, and ii) treating both  $t2g$  and  $eg$  with DMFT. In the first case, the valences are similar in all three double-counting formulas. The  $t2g$  occupancy is very close to nominal value 2. The exact double-counting is again smaller than given by FLL or nominal formula, which leads to a slightly larger splitting between oxygen- $p$  and  $V-d$  states, i.e., slight upward shift of oxygen states in Fig. 2a. In case ii) displayed in Fig. 2b and tabulated in table IV, where both the  $t2g$  and  $eg$  orbitals are treated by DMFT, the FLL formula dramatically fails, as it overestimates the valence, i.e.,  $n_d^{FLL} - n_d^{exact} \approx 0.26$ . While the Mott gap does not entirely collapse, it is severely underestimated by FLL formula. The nominal valence, however, gives very similar results as the exact DC. This improvement of nominal DC as compared to FLL was pointed out in Refs. 9, 10, and was found to hold not just in transition metal oxides but also in actinides [21]. The  $t2g$  occupancy  $n_{t2g}$  in the nominal and exact DC is very close to nominal value of 2, equal to the scheme i) presented above. It is therefore not surprising that the spectra in Fig. 2a, and Fig. 2b are similar, with slight improvement compared to experiment when  $eg$  orbitals are also treated by DMFT.

In summary, we presented continuum representation of the Dynamical Mean Field Theory, which allowed us to derive an exact double-counting between Dynamical Mean Field Theory and Density Functional Theory. The implementation of exact double-counting for solids shows the improved agreement with experiment as compared to standard FLL formula. Previously introduced nominal DC formula [9, 10] is in very good agreement with exact double-counting derived here.

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